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Recycle of Modified Fly Ash from Furnace Sorbent Injection

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RECYCLE OF MODIFIED FLY ASH FROM FURNACE SORBENT INJECTION

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16. ABSTRACT The paper discusses technical and economic studies to assess the impact of recycle on the furnace sorbent injection process. Levelized costs of various recycle schemes were compared to baseline (non-recycle) costs using the EPA LIMB Cost Model and the LIMB Recycle Model. Laboratory and pilot-scale tests were performed to evaluate the technical feasibility of the recycle schemes. Use of recycle to enable replacement of expensive hydrated lime sorbent with limestone suggested possible savings of 21% in 15-year levelized costs per ton of SO2 removed. This process may not be feasible, however, due to sorbent disintegration. Recycle with quicklime and hydrated lime as sorbents gave projected savings of up to $79 and $127 per ton of SO2 removed, respectively. Pilot-scale studies showed that 78-90% hydration of unused sorbent could be achieved in 30-263 minutes. Of the separation processes investigated, only cyclonic separation and steam elutriation were found to be feasible.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS		b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group	
Pollution	Calcium Oxides	Pollution Control	13B	
Furnaces	Cyclone Separators	Stationary Sources	13A	07A, 13I
Sulfur Dioxide	Elutriation	Furnace Sorbent Injection	07B	07D
Sorbents			11G	
Fly Ash		Recycling	21B	
Limestone		Hydrated Lime	08G	

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INTRODUCTION

Description of Furnace Sorbent Injection Process

Furnace sorbent injection is being developed as a low-cost alternative to achieve moderate degrees of SO_2 control at coal-fired power plants. In this process, a calcium-based sorbent is injected into the upper furnace, where the temperature is typically about 2200°F (1200°C). The sorbent may be limestone ($CaCO_3$), quicklime (CaO), or hydrated lime [$Ca(OH)_2$]. With either limestone or hydrated lime, rapid decomposition takes place, yielding CaO.

$$CaCO_3 \quad ----> \quad CaO \quad + \quad CO_2$$

$$Ca(OH)_2 \quad ----> \quad CaO \quad + \quad H_2O$$

The CaO then reacts with SO_2 and O_2 to form calcium sulfate ($CaSO_4$).

$$CaO \quad + \quad SO_2 \quad + \quad 1/2O_2 \quad ----> \quad CaSO_4$$

Generally, only about 10 to 30% of the CaO is converted to $CaSO_4$, with the CaO produced from hydrated lime being most reactive, and the CaO produced from limestone being least reactive. In any case, no more than about 30% of the calcium content is utilized (i.e., reacted with SO_2).

Incentive for Recovering and Recycling Unused Sorbent

Since sorbent utilization is so low in furnace sorbent injection, an excess amount of sorbent is required to achieve moderate SO_2 removals. For example, twice the stoichiometric amount of sorbent is typically required to achieve 60% removal with hydrated lime. Even higher amounts would be required with less effective sorbents. This means that at least 70% of the sorbent calcium content is wasted and collected along with the coal fly ash as unreacted CaO. There is clearly a need to reduce this waste by increasing sorbent utilization. One way of accomplishing this is by recycle.

By using a recycle process in conjunction with furnace sorbent injection, the overall utilization of the sorbent can be increased. This would allow the user to reduce the consumption of fresh sorbent, while also reducing the volume of waste products generated. This should make furnace sorbent injection more attractive economically, if the cost savings are greater than the cost of equipment required for recycle.

In some cases, recycle may also allow the use of cheaper sorbents. For example, an inexpensive coarse limestone may be used with recycle to replace a once-through process using the more expensive hydrated lime. Alternatively, quicklime may be used and hydrated on site along with the unreacted CaO in the recycled material.

Conceptual Processes for Sorbent Recovery and Recycle

Potential recycle processes differ with respect to feedstock and method of ash/sorbent separation. In general, separation is always required to avoid excessive particulate loadings in the boiler and dust collectors. Figure 1 shows the generalized conceptual process. As shown, the feedstock may be limestone, hydrated lime, or quicklime. The latter must be hydrated on site prior to injection, since commercial quicklime is generally not sufficiently reactive.

With a coarse limestone feedstock, the resulting large particles of unreacted sorbent may be best collected ahead of the existing particulate collector, using a cyclone or multiclone. Alternatively, the inlet field of an electrostatic precipitator (ESP) may be used to collect the coarse limestone sorbent, since it will be much coarser than the fly ash.

With quicklime and hydrated lime, the ash/sorbent mixture may be collected in the existing ESP or baghouse. Dense-phase separation can then be used to remove the relatively large fly ash particles, which have a mass median diameter (mmd) of about 15 to 20 μm. The sorbent particles typically have a mmd of 2 to 4 μm. This type of separation can be achieved using equipment similar to the size classifiers at commercial lime hydration plants. The separated sorbent can then be hydrated to convert the CaO to its more reactive form. The ash from quicklime injection could be hydrated in the same vessel used to hydrate the fresh quicklime feedstock. This study, however, assumes that two separate hydrators are used. This enhances system reliability by providing some equipment redundancy.

Objectives of This Study

The purpose of this study is to determine the technical and economic feasibility of recycle as a means of improving the furnace sorbent injection process. Technically, this means:

1. Determining if the unused sorbent can be reactivated by hydration,

2. Evaluating the effectiveness of the reactivated sorbent by reinjection into a furnace, and

3. Identifying and evaluating methods of separating the coal fly ash from the sorbent.

Economically, the key objectives are:

1. To compare the costs of various recycle concepts with those of a baseline (non-recycle) furnace sorbent injection process,

2. To identify the most attractive recycle strategy, and

3. To guide the recycle testing by concentrating efforts on the processes with the largest potential payoff.

ECONOMIC EVALUATIONS

Approach

The economic evaluations of recycle were based on the use of modified versions of EPA's LIMB (Limestone Injection Multistage Burner) Cost Model and Recycle Model (1). The LIMB Cost Model, which is written for the non-recycle case only, was modified to include an on-site hydrator when quicklime is specified as the feedstock. The Recycle Model, which was originally written for the ideal case of perfect ash/sorbent separation, was modified to allow for various degrees of non-ideal separation. The entire system material balance and equipment sizing was modified accordingly. Equations for sizing and costing the recycle separator/hydrator system were also incorporated into the model. Equations for the capital costs were obtained by scaling quotes obtained from a major supplier of lime hydrator/classifier systems. The quotes included all of the required peripheral equipment, such as conveyors and pneumatic reinjection equipment.

With the Recycle Model modified to include all of the required recycle equipment, it was used to predict levelized costs for various recycle processes using coarse limestone, quicklime, and hydrated lime feedstocks. For comparison, the modified LIMB Cost Model was used to predict the levelized costs of baseline (non-recycle) processes using the same feedstocks and achieving the same levels of SO_2 removal. The parameters used in these economic analyses are summarized in Table 1. These parameters represent average values for all power plants east of the Mississippi River, which includes most plants that are likely candidates for furnace sorbent injection retrofits.

Coarse Limestone Injection with Cyclonic Separation

Only a preliminary economic evaluation was made for this concept. Detailed economic studies were not performed since pilot testing revealed that the coarse limestone disintegrated, making it impossible to achieve the desired ash/sorbent separation. The economic results will be briefly discussed here in case other research identifies coarse limestones that do not disintegrate.

Use of a coarse limestone feedstock at twice the stoichiometric rate was compared to non-recycle hydrated lime injection at the same stoichiometry. An overall SO_2 removal of 50% was assumed for both. Limestone injection without recycle was not evaluated since it was believed that 50% removal could not be achieved with limestone without recycle. Based on previous test results, limestone and hydrated lime utilizations were assumed to be 10% and 25%, respectively. For recycle, this fixed the required recycle ratio at 44.5%.

Based on that figure, material balances were performed and all equipment was sized.

The separator/hydrator system mentioned above was reduced to a hydrator system alone for this case, and a separate cyclonic separator was sized and costed. This approach was deemed most appropriate for the coarse sorbent, which apparently could be readily separated from the smaller fly ash particles using a cyclonic precollector. The capital cost of the cyclone was based on another vendor quote, and a booster fan to compensate for the cyclone pressure drop was also costed. The operating cost of the booster fan was also included based on the pressure drop and flue gas flowrate. A pressure drop of 2 in. (5 cm) of water was assumed.

The comparison of the two systems described above showed that recycle offered a considerable cost savings. This was primarily attributable to the much lower cost of the coarse limestone [assumed to be $18 per ton ($20 per metric tonne) delivered, versus $71 per ton ($78 per metric tonne) for hydrated lime]. This was true even though it was assumed that recycle would require an additional operator per shift (in addition to two additional operators required for the furnace sorbent injection itself). The savings in sorbent costs were more than sufficient to offset the costs of the recycle equipment, cyclone, booster fan, additional operation and maintenance, and increased use of in-plant power. The net result for the two scenarios compared was a 21% saving in 15-year levelized cost per ton of SO_2 removed.

For the two cases described above, recycle resulted in a 31% increase in the solids loading in the boiler, assuming that the cyclone collects only the coarse sorbent and no fly ash. This idealized case may be approached if coarse (e.g., mmd > 80 μm) limestone is used, and it does not disintegrate in the boiler. As described later in this paper, experimental work has shown that disintegration is a problem, at least with the particular limestone tested. However, it is possible that other limestones may not exhibit this behavior. Thus, the coarse limestone recycle concept may still offer significant potential for improving the furnace sorbent injection process.

Hydrated Lime Injection with Separation and Hydration of Recycled Sorbent

A more thorough economic analysis was performed for this scenario. Again, a recycle hydrator was sized and costed by vendor quotation. For this analysis, a dense-phase separator, which removes most of the larger fly ash particles from the smaller sorbent particles, was included ahead of the hydrator. For costing purposes, the separator was assumed to be similar to the classifiers used on commercial lime hydration systems. To allow for various feedrates, the vendor cost quotation was scaled using a 0.6 exponent on the input flowrate to the separator/hydrator system. Detailed material balances were performed in the model to allow the simulation of various degrees of ash/sorbent separation.

For the cases described here, it was assumed that 75% of the ash/sorbent mixture collected in the plant dust collectors was recycled to the separator/hydrator system. It was further assumed that 80% of the recycled sorbent

was carried through the separator (i.e., 20% was lost along with the fly ash). This corresponds to an effective sorbent recycle ratio of 60%.

The baseline (non-recycle) analysis was compared to the above recycle analysis assuming that hydrated lime was used as the sorbent feedstock in both. Comparisons were made at three different sorbent costs: $50, $60, and $70 per ton ($55, $66, and $77 per metric tonne) of hydrated lime delivered. The effect of different operating requirements was also examined by varying the number of additional operators required. Two additional operators were specified for the baseline scheme, and three or four additional operators were specified for the recycle scheme (i.e., one or two operators in addition to the requirements of a baseline furnace injection system).

The results of this comparison are summarized in Table 2. Despite the higher operating requirements, recycle still offers an economic advantage over the baseline furnace injection system. For the scenarios examined, savings as large as $127 per ton ($140 per metric tonne) of SO_2 removed can be realized. These are expressed in terms of 15-year levelized costs. Based on these results, there is clearly a strong incentive to consider recycle with hydrate-based systems. Of course, this incentive is reduced as the sorbent cost decreases, since this is the primary source of cost savings.

Quicklime Hydration and Injection with Ash Separation and Hydration

This comparison was made in a manner analogous to the hydrate-based scenarios discussed above. However, a second hydrator was added to provide for separate hydration of the fresh quicklime and the recycled sorbent. This equipment redundancy allows system operation in the non-recycle mode if necessary. Again, it was assumed that the ash/sorbent separation was carried out in the dense phase ahead of the recycle hydrator. The same assumptions were made concerning the fraction of ash/sorbent mixture recycled and the fraction of sorbent carried through the separator. Again, the effects of various sorbent costs and operating requirements were also examined. In this case, however, lower sorbent costs were assumed for the less expensive quicklime [$40, $50, and $60 per ton ($44, $55, and $66 per metric tonne) of quicklime delivered].

The results for the quicklime-based systems are summarized in Table 3. While the savings are less than with hydrate-based systems, it is also apparent that the absolute costs are lower using quicklime. This is due to the lower unit cost of the sorbent and the lower tonnage requirements resulting from the lower molecular weight. In fact, it appears that a quicklime-based recycle system offers the lowest potential levelized cost [as low as $685 per ton ($754 per metric tonne) of SO_2 removed at the lowest sorbent cost and minimum operating requirements]. Even at the highest sorbent cost and operating requirement, the quicklime-based recycle system appears to be more cost-effective than the cheapest hydrate-based recycle system. This suggests that future development of the furnace injection process should be directed toward quicklime-based systems with recycle.

LABORATORY AND PILOT FEASIBILITY STUDIES

Previous work has shown that calcium-based furnace sorbent injection ash can be reactivated by hydration and reinjected to obtain SO_2 capture that is only 5-10% lower than that obtained by hydrated lime (2,3,4). These findings prompted efforts to separate the unused sorbent from the fly ash, and optimize the hydration process. Results of these investigations are discussed below.

Laboratory Hydration Studies Using Coarse Limestone Ash

Parameters affecting completeness and rate of the hydration reaction were studied using a 4.5-qt (4.25-L) KitchenAid mixer. Parameters investigated included water temperature, water-to-lime molar ratio, and use of wetting and dispersion agents. Results of these efforts indicated that variations in water temperature and use of chemical additives did not produce any additional hydration.

Water-to-lime ratios were found to significantly affect the rate and degree of hydration. This is shown in Figure 2. The results indicated: (1) more than 80% hydration can be achieved within 4-5 hrs with molar water-to-lime ratios of 4-9, and (2) at the lower water-to-lime ratios (less than 4) there was some dependency of the degree of hydration on the hydration time.

Hydration studies were also performed using ash from hydrated lime injection. These studies, performed in the laboratory and on a pilot scale, indicated high degrees of hydration within short periods of time. Slaking tests showed a medium-to-low reactivity level in accordance with ASTM C110. This indicates that the slaking rate is low relative to quicklime, but the time required for ash hydration is still reasonable (<30 min).

Laboratory Separation Studies Using Raw and Hydrated Ash

Separation of sorbent from the fly ash is necessary to avoid excessive particulate loading of the gases entering the boiler and the particulate collection systems. Several potential separation techniques have been proposed including: (1) use of an ESP; (2) inertial or cyclonic separation; (3) steam hydration/elutriation; (4) magnetic separation; (5) froth flotation; (6) dense media separation; and (7) liquid extraction. Dry separation is generally preferred over wet separation since it avoids potential problems with water pollution and slurry containment. Use of an ESP, magnetic separation, and froth flotation have been described in previous work (3).

Cyclonic Separation of Ash/Sorbent Mixtures. Two different separation schemes have been proposed which take advantage of the fact that the mean fly ash particle size is approximately 20 μm. Use of hydrated lime with mean particle diameter of 2 to 5 μm would enable separation from the larger fly ash particles. Previous work (4) indicated that a cutpoint of around 2 μm could be used to remove most of the fly ash from the waste product.

Another separation and recovery scheme involves the use of coarse limestone particles with mean particle sizes larger than 80 μm. After calcination, these particles should be larger than 60 μm, so they can be separated from the much smaller fly ash particles by a cyclone. This material could then be hydrated and injected back into the furnace as hydrated lime. Results obtained with this separation technique are discussed under pilot-scale studies.

Air/Steam Elutriation. Air/steam elutriation is still in the laboratory stage. In the steam process, a bed of sorbent/ash particles are hydrated and separated using an upward flow of steam. The smaller hydrate particles are entrained in the steam flow and carried out of the bed. The laboratory apparatus used for studying this process has been described previously (4). Steam carrying the hydrated particles could, in a full-scale process, be blown directly into the boiler for flue gas desulfurization. Results of preliminary tests show good hydration, but low hydrate carryover efficiencies have been obtained. The ash can be hydrated to 85% hydration with the steam, but once hydrated, the particles agglomerate in the reaction vessel, for both the coarse limestone ash and the hydrated lime ash. Vibration was found to deter agglomeration. Fluidization was also better with vibration, but still no significant amount of hydrate particles were elutriated.

Experimental results indicate that: (1) at below 130°C, the degree of hydration is inversely proportional to reaction flask temperature (see Figure 3), and (2) higher steam flow rates result in more carryover with less separation (see Figure 4). The inverse relationship between degree of hydration and reaction flask temperature below 130°C is due to the conditions required to hydrate the sulfate shell. Hydrated forms of calcium sulfate are not stable above 130°C.

Air elutriation has not yet been tested. For coarse limestone ash, the smaller fly ash particles would be elutriated prior to hydration. For the hydrated lime ash, the mixture would first be hydrated, and the smaller hydrate particles would then be elutriated.

Dense Media Separation. This method of recovering unspent sorbent makes use of the change in density which accompanies the reaction of CaO (sp gr = 3.3) with water to form $Ca(OH)_2$ (sp gr = 2.2). The process involves mixing the ash with a dense medium (e.g., tetrabromoethane, sp gr = 2.97) and allowing it to separate into two fractions. The fraction which floats will have a density less than 2.97, while the calcined sorbent (CaO) and other compounds having a density greater than 2.97 will sink. The heavy fraction containing the CaO is then mixed with water to hydrate the CaO to $Ca(OH)_2$. The hydrated sorbent particles (sp gr = 2.2) can then be separated from the remaining heavier fraction by repeating the process. After the first separation, the light fraction contained 60.7% CaO, while the heavy fraction contained 49.3% by weight. Further testing of this concept was not deemed feasible due to the prohibitive length of time required to achieve separation.

Liquid Extraction. This process takes advantage of the fact that the solubility of lime in water increases dramatically with addition of sugar,

and declines with increased water temperature. Coarse limestone injection ash was added to a 50% saturated sugar solution. After mixing, the liquid was decanted and heated to precipitate the $Ca(OH)_2$. Solids collected after filtration were found to be 95% CaO on an ignited basis. Despite this high purity product, low recovery and filtration problems prevented this method from being feasible.

Pilot-Scale Hydration Studies Using Recycled Ash

Pilot-scale hydration was studied using ash produced from fine limestone, coarse limestone, and hydrated lime injection. The results obtained with the fine limestone and hydrated lime ash have been discussed previously (3,4). Therefore, only the results obtained from the coarse limestone ash are reported here. These studies were done using a 1-ft^3 (28-L) ribbon blender. Using a water-to-lime ratio of 4:1 and hydration times of 30 to 263 min, the percent hydration varied from 78 to 90%. These experiments proved that the laboratory results could be successfully scaled up. The higher degree of hydration achieved in the larger scale tests is attributed to more uniform mixing and better contact between the water and ash particles due to the use of more efficient mixing.

Pilot Separation Studies Using Raw and Hydrated Ash

The Southern Research Institute (SRI) combustor outlet was equipped with a funnel-type divertor to channel gas flow into the pilot-scale cyclone shown in Figure 5. Three 6-ft (1.8-m) water-cooled heat exchanger sections were installed between the divertor and the cyclone to cool the flue gas below 400°F (200°C) and prevent damage to the Teflon seals on the cyclone.

Initial tests were made firing coal without sorbent injection. Mass train samples were collected at both the cyclone inlet and outlet. Analysis of these samples showed that approximately 70% of the fly ash was collected in the cyclone. From this result, it was clear that the cyclone cutpoint was too small. Gas flow reduction was not feasible as the combustor was being operated at minimum load. Discussions with the cyclone manufacturer indicated that modifications to the cyclone would be difficult and had little likelihood of producing satisfactory results.

While the cyclone performance was unsatisfactory for particle separation, runs were conducted with limestone injection to determine if particle disintegration was a problem. Tenn-Luttrell limestone (mmd = 80 μm) was injected into the combustor under both hot (natural-gas-fired) and cold (air flow only) gas flow conditions. Particle size distributions were measured at the cyclone inlet. Sample analysis indicated that disintegration did not occur during cold flow testing. However, severe disintegration occurred during hot flow testing as the mmd of the limestone particles decreased by a factor of 7 or 8. Based on these results, the coarse limestone recycle concept does not appear feasible, unless a different limestone, which does not disintegrate, can be identified.

CONCLUSIONS

Economic Evaluations

Based on the economic evaluations discussed above, significant cost savings can be realized by use of a recycle process in conjunction with furnace sorbent injection. The largest savings appear to result from the replacement of an expensive sorbent with a less costly feedstock (e.g., replacing hydrated lime with coarse limestone). However, recycle may not be feasible with coarse limestone injection due to sorbent disintegration, which makes it impossible to separate the ash and sorbent. Further work is needed to determine if this is a general problem with all limestone sources. If so, a quicklime-based recycle system appears to offer the lowest overall cost. However, significant cost savings are possible with both the hydrate- and quicklime-based systems. The actual savings realized are strongly dependent on the sorbent cost and the additional operating requirements of the recycle system.

Feasibility Studies

Unused sorbent can be reactivated and reinjected to obtain additional SO_2 capture. SO_2 removal by the reactivated sorbent (without separation) was found to be slightly lower than that obtained with hydrated lime for similar Ca/S ratios, but approximately the same for comparable $Ca(OH)_2$/S ratios. Pilot-scale studies indicated that 78 to 90% of the unused sorbent could be hydrated within a reasonable time.

Minimal separation of unreacted sorbent was possible by cyclonic separation. Disintegration of coarse limestone particles prevented significant separation. Dense media separation and liquid extraction do not appear to be feasible. Steam elutriation, while still in the laboratory development phase, appears to have good potential.

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4. R. S. Dahlin, C. L. Lishawa, and N. Kaplan, "Analysis of LIMB Waste Management Options," in Proceedings: 1986 Joint Symposium on Dry SO_2 and Simultaneous SO_2/NO_x Control Technologies, Vol. 2. EPA-600/9-86-029b (NTIS PB87-120457), October 1986.

Table 1. PARAMETERS USED IN ECONOMIC EVALUATION

Coal Sulfur Content - 2.2%	Coal Ash Content - 15.1%
Heating Value - 11,900 Btu/lb (6.61 kcal/g)	Heat Rate - 9800 Btu/kWhr (2470 kcal/kWhr)
Capacity Factor - 50%	Load - 300 MW
Ca/S Ratio - 2:1	SO_2 Removal - 50%
Book Life - 15 years	Tax Life - 5 years
Labor Rate - $21.00/hr	Waste Disposal Cost - $7.66/ton ($8.43/metric tonne)
In-Plant Power Cost - 54 mills/kWhr	Status - Retrofit

Table 2. RESULTS OF ECONOMIC COMPARISONS FOR HYDRATE-BASED SYSTEMS

Sorbent Cost, $/ton[a]	No. of Operating Jobs[b]	Levelized Cost, $/ton SO_2 Removed[a] Baseline	Recycle	Cost Savings
50	2/3	853	792	61
50	2/4	853	822	31
60	2/3	931	834	97
60	2/4	931	864	67
70	2/3	1004	877	127
70	2/4	1004	906	98

a. $/metric tonne = $/ton x 1.1.
b. No. of additional operators without recycle/No. of additional operators with recycle.

Table 3. RESULTS OF ECONOMIC COMPARISONS FOR QUICKLIME-BASED SYSTEMS

Sorbent Cost, $/ton[a]	No. of Operating Jobs[b]	Levelized Cost, $/ton SO_2 Removed[a]		
		Baseline	Recycle	Cost Savings
40	2/3	717	685	32
40	2/4	717	715	2
50	2/3	772	717	55
50	2/4	772	746	26
60	2/3	827	748	79
60	2/4	827	778	49

a. $/metric tonne = $/ton x 1.1.
b. No. of additional operators without recycle/No. of additional operators with recycle.

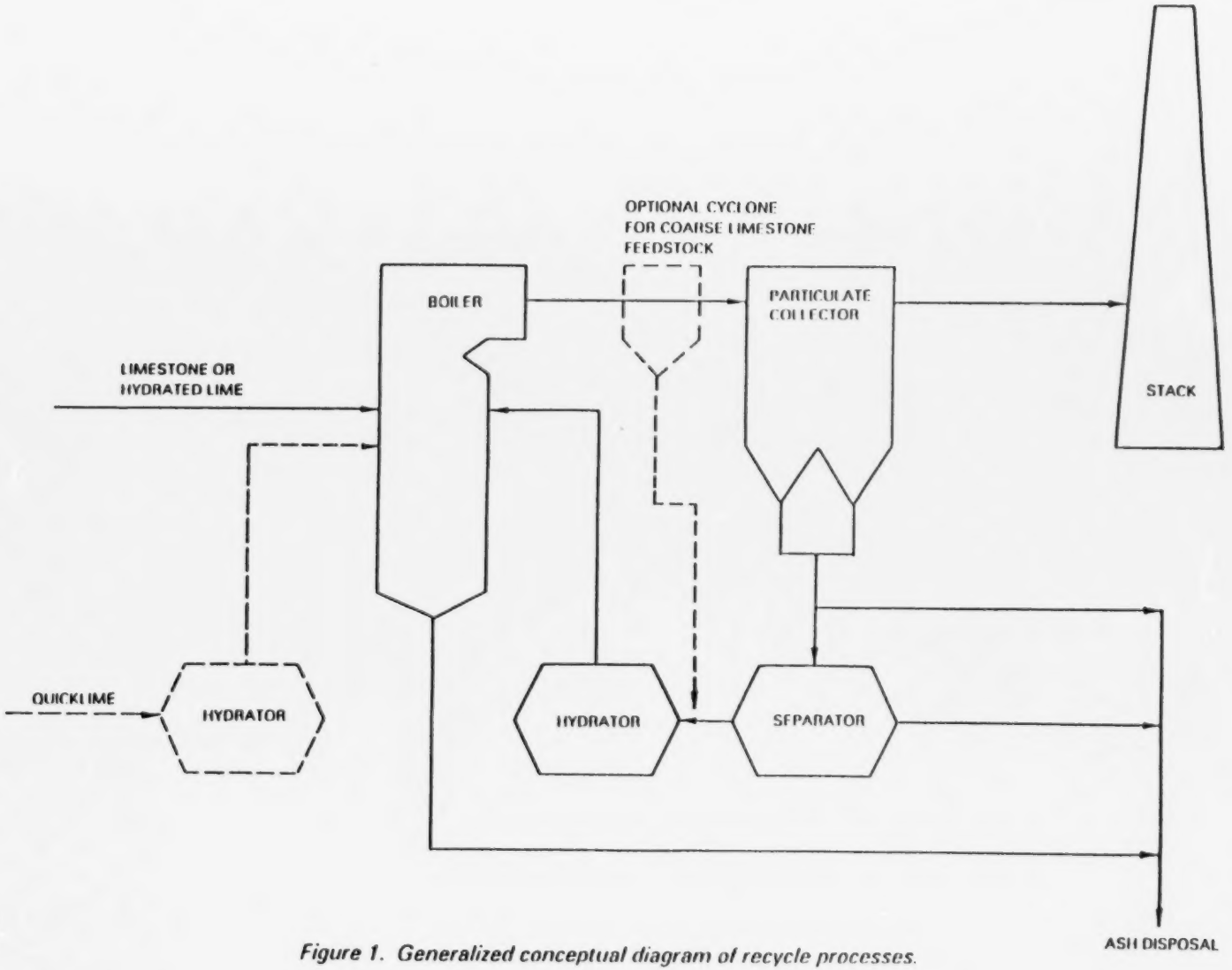

Figure 1. Generalized conceptual diagram of recycle processes.

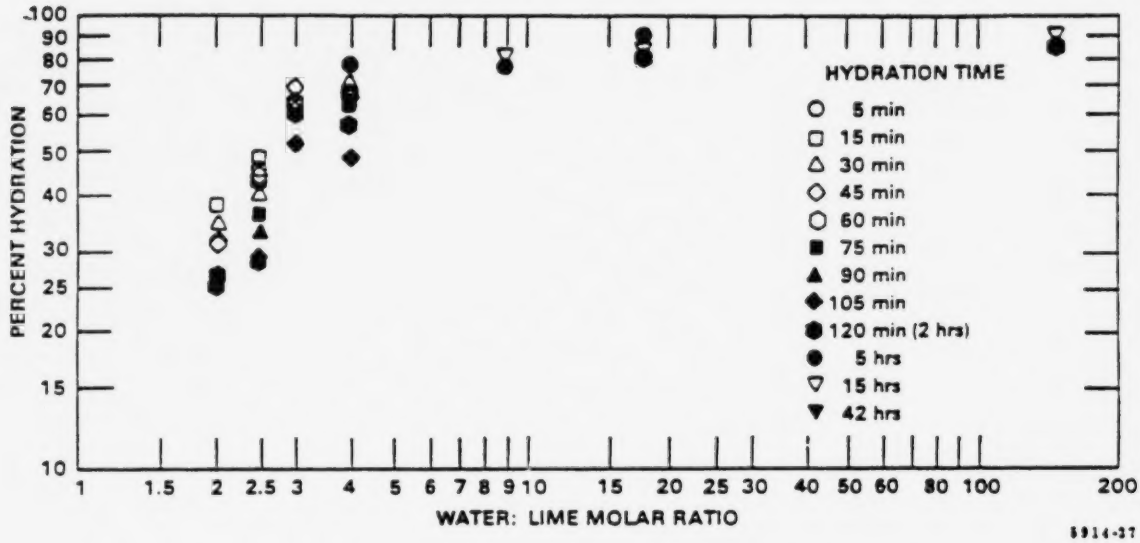

Figure 2. Effect of water-to-lime ratio and hydration time on percent hydration.

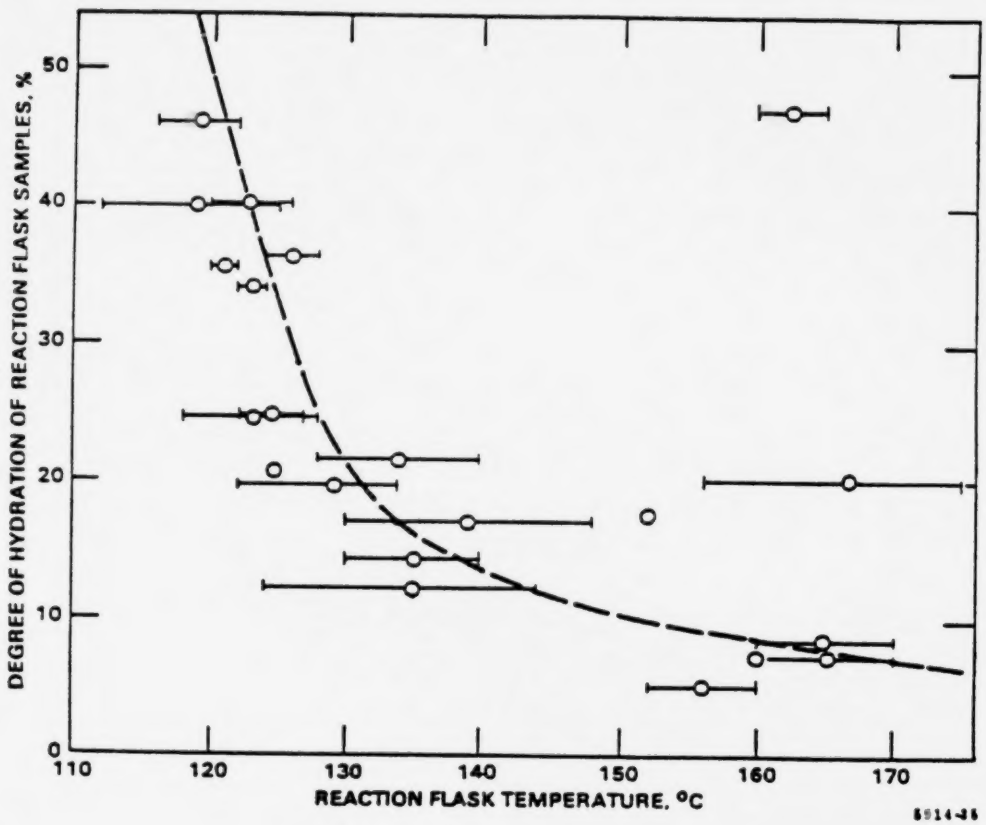

Figure 3. Effect of reaction flask temperature on degree of hydration.

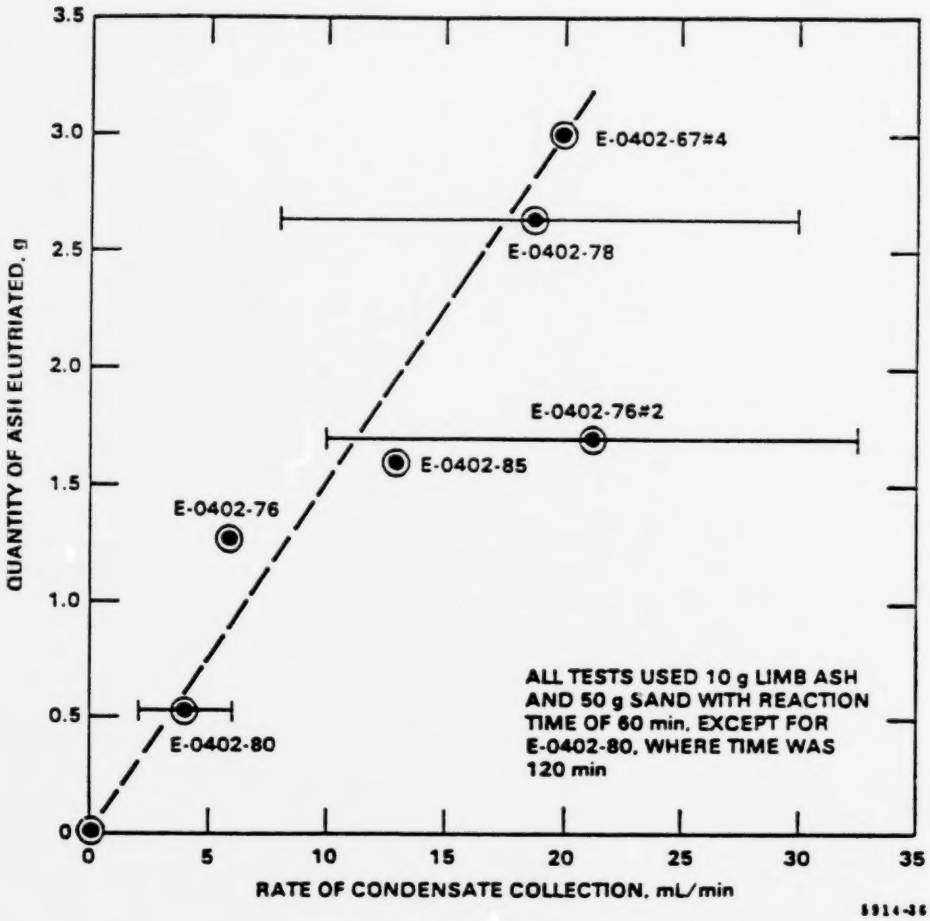

Figure 4. Effect of steam flow on carryover.

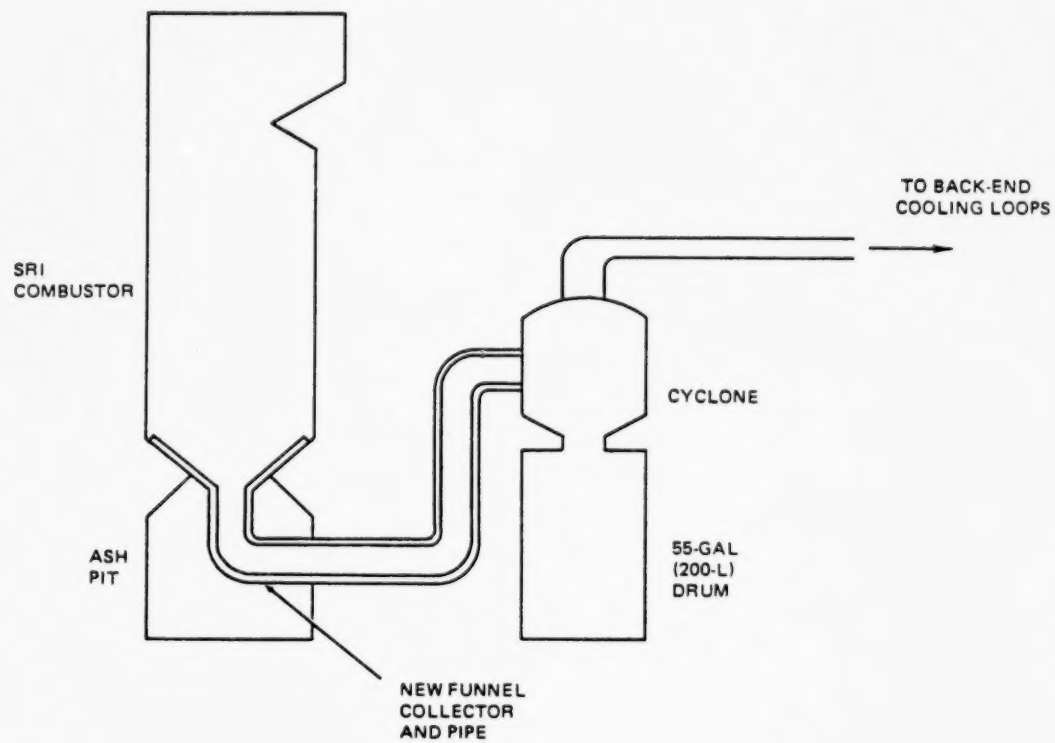

Figure 5. Cyclone set up.